Journal of the Korean Ceramic Society
Vol. 45, No. 4, pp. 204~207, 2008.

Fabrication of SiC Fiber-SiC Matrix Composites by Reaction Sintering

Kwang-Young Lim, Young-Wook Kim†, and Ji Yeon Park*

Department of Materials Science and Engineering, University of Seoul, Seoul 130-743, Korea
**Division of Nuclear Materials Technology Development, Korea Atomic Energy Research Institute, Daejon 305-353, Korea*
(Received March 31, 2008; Accepted April 8, 2008)

ABSTRACT

This paper presents a new process for producing SiC fiber-SiC matrix (SiC/SiC) composites by reaction sintering. The processing strategy for the fabrication of the SiC/SiC composites involves the following: (1) infiltration of the SiC fiber fabric using a slurry consisting of Si and C precursors, (2) stacking the slurry-infiltrated SiC fiber fabric at room temperature, (3) cross-linking the stacked composites, (4) pyrolysis of the stacked composites, and (5) hot-pressing of the pyrolyzed composites. It was possible to obtain dense SiC/SiC composites with relative densities of >96% and a typical flexural strength of ~400 MPa.

Key words : Fiber composites, Silicon carbide, Reaction sintering, Strength

1. Introduction

Silicon carbide fiber-reinforced SiC matrix composites (SiC/SiC composites) are promising candidate materials for high-temperature structural applications and fusion power reactor applications. Applications of the former type include gas turbine hot section parts, and applications of the latter type include the fabrication of the first wall, blanket, and diverter of nuclear fusion reactors, which require high thermal conductivity, excellent hermeticity with respect to gases, and low residual radioactivity.[1-3] Typical fabrication processes for SiC/SiC composites are chemical vapor infiltration (CVI),[4-6] precursor impregnation and pyrolysis (PIP),[7,8] slurry infiltration,[9,10] and hybrid processes of the above routes, e.g., a few PIP sequences with a polymer precursor and a deposition of SiC from the gas phase by CVI.[11,12] With regard to protection of fibers and interfaces from the oxidation environment during application, a dense matrix is preferable. In terms of densification, the CVI and PIP processes and a hybrid of those processes yield composites with significant residual porosity.

In our previous works,[13,14] a new process based on stacking infiltrated SiC fiber fabric and SiC tapes alternately to fabricate SiC/SiC composites was developed. The developed process produced dense (>98% of theoretical density) 2D SiC/SiC composites. Another process developed was a process based on slurry infiltration and a stacking process. This process also produced dense (>98% of theoretical density) 2D SiC/SiC composites. In this work, we investigated a reaction sintering process based on Si- and C-precursors infiltration and a stacking process to fabricate dense SiC/SiC composites.

†Corresponding author : Young-Wook Kim
E-mail : ywkim@uos.ac.kr
Tel : +82-2-2210-2760 Fax : +82-2-2215-5863

2. Experimental Procedure

2D woven Tyranno SA fiber fabrics (SiC fiber fabrics, Ube Industries Ltd., Japan) were used as the reinforcement for the fabrication of SiC/SiC composites. Fig. 1 shows the steps of the procedure for fabricating dense SiC fiber-SiC matrix composites. Nonaqueous slurry was prepared by mixing 59.4 wt% Si source (polysiloxane, GE Toshiba Silicones Co., Ltd., Tokyo, Japan), 30.6 wt% carbon source (phenol resin, Kangnam Chemical Co., Ltd., Incheon, Korea), 6.4 wt% Al_2O_3, 2.6 wt% Y_2O_3, and 1 wt% MgO for 1 h in a Teflon bottle using SiC grinding media. The Al_2O_3-Y_2O_3-MgO mixture was added as a sintering additive. The fiber fabric was cut into a rectangular shape (21 mm×25 mm) and infiltrated in the slurry using an ultrasonic bath, and then dried at room temperature. The slurry-infiltrated fiber fabric was stacked at room temperature under a pressure of 10 MPa. The stacked sheets were cross-linked at 200°C, and then pyrolyzed at 800°C for 1 h under a flow of argon to remove the organic components. The pyrolyzed compact was hot-pressed at 1750 °C for 1-4 h under a pressure of 15 MPa in an argon atmosphere.

The density of each sample was measured using the Archimedes method. The hot-pressed specimens were cut into 3×2.5×25 mm samples for strength measurements. The bend test was performed at room temperature on six specimens for each condition using a four-point method with inner and outer spans of 10 and 20 mm, respectively. The fracture surfaces were observed via scanning electron microscopy (SEM).

3. Results and Discussion

The pyrolysis process at 800°C converted the precursors into amorphous SiOC and C, and subsequent heating up to

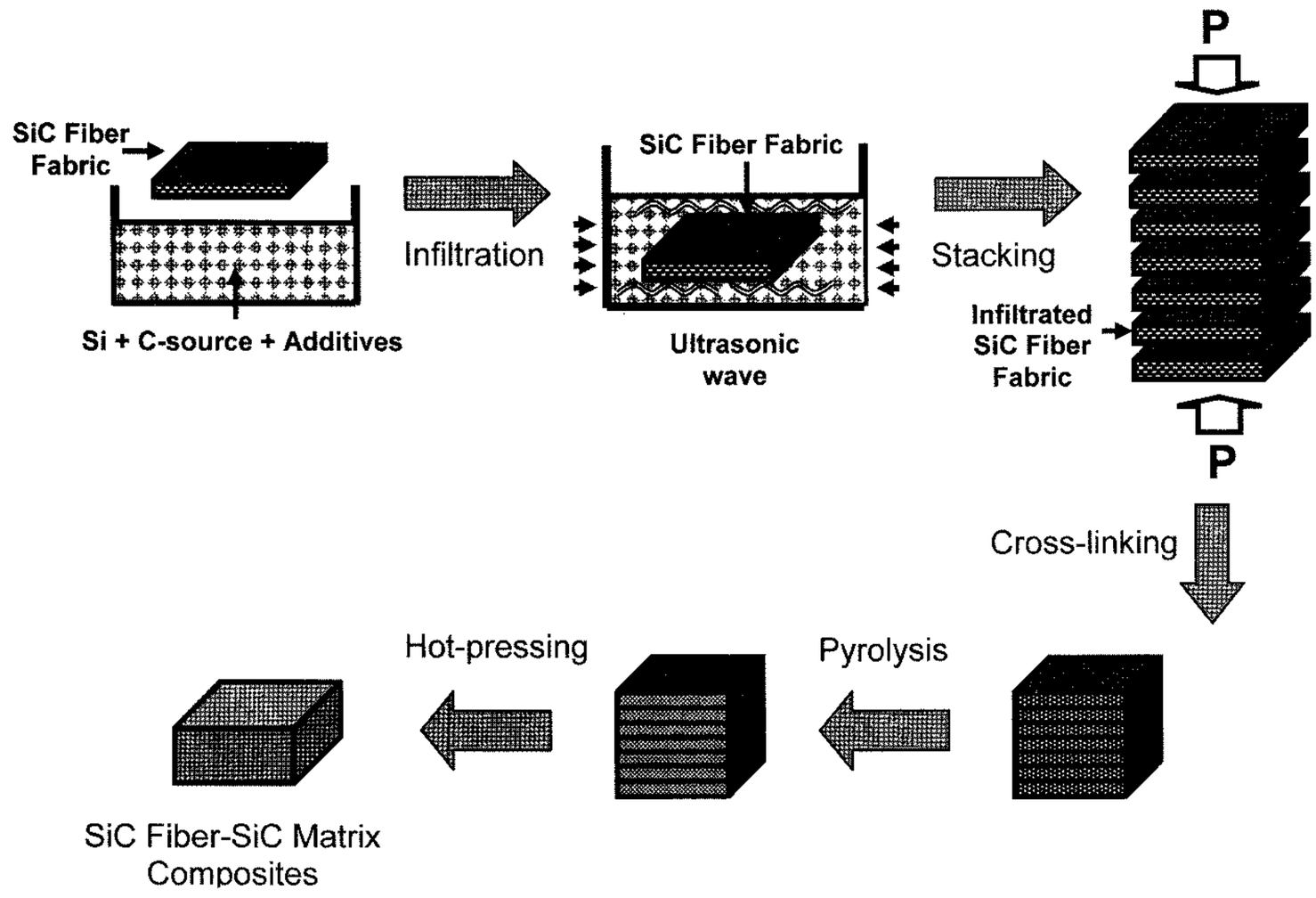

Fig. 1. Steps in the procedure for fabricating SiC fiber-SiC matrix composites by reaction sintering.

1750°C led to the *in situ* synthesis of SiC via the reaction between SiOC and C.[15,16] During hot-pressing, the additives formed a liquid, and the synthesized SiC was sintered via liquid-phase sintering.

Fig. 2 shows the variations in the density of the synthesized SiC/SiC composites under different sintering times. The sintered density increased from 3.03 g/cm^3 to 3.04 g/cm^3 with increasing sintering time from 1 h to 2 h. Increasing the sintering time to 4 h at 1750°C led to a density of 3.06 g/cm^3. The densification process for the composites was divided into two main parts: matrix densification and inter-fiber densification. Densification of the matrix is similar to that for monolithic SiC and is a liquid-phase sintering and temperature-dependent process. From a previous study on the densification of monolithic SiC,[17] a highly-densified matrix should be achieved at 1750°C for the additive composition, even though fiber inclusion slows the densification process.[10,13] Inter-fiber densification may be controlled by the deformation of fibers during hot pressing. This will be discussed later.

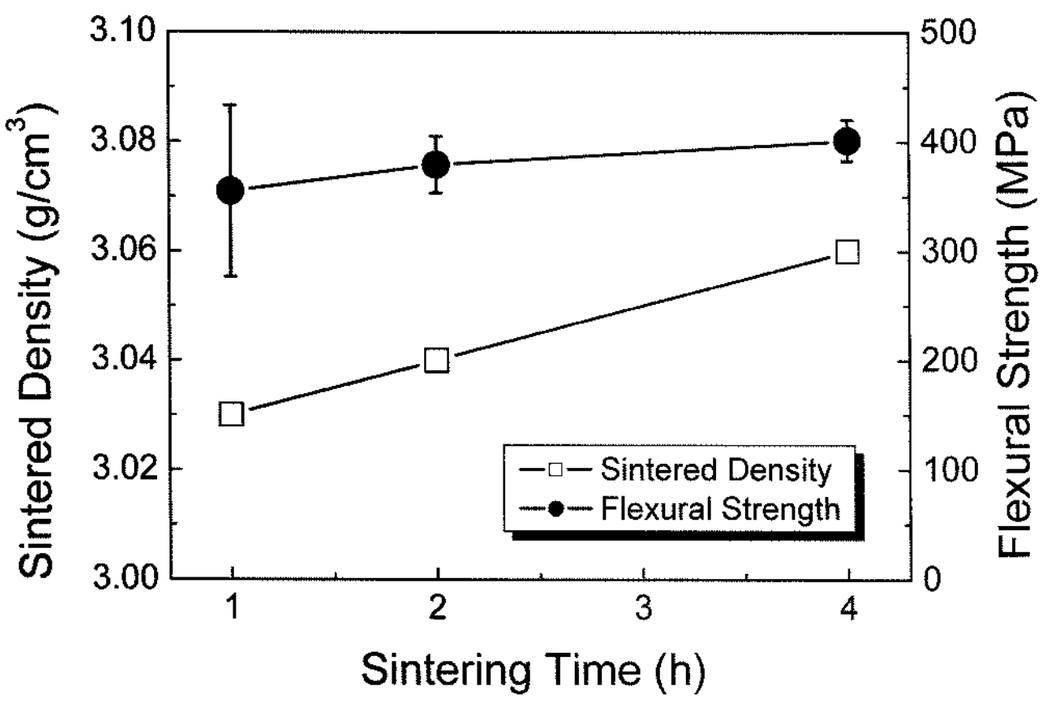

Fig. 2. Effect of sintering time on the densification and strength of the SiC fiber-SiC matrix composites.

The sintered density of the SiC/SiC composites fabricated by the NITE process ranged from 2.77 g/cm^3 to 2.93 g/cm^3.[9,10] Therefore, the densities obtained (3.03~3.06 g/cm^3) in this study were higher than those of the NITE-processed SiC/SiC composites. This suggests that the present new process consisting of the precursor slurry-infiltration and stacking process is an efficient way for fabricating dense SiC/SiC composites.

Fig. 3 shows the microstructure of the fracture surfaces for the composites sintered at 1750°C for various times. Under the present fabrication method, the distinction between the SiC fiber bundle and SiC matrix in the composites was quite clear up to 2 h. However, further increasing the sintering time to 4 h at 1750°C led to unclear distinction between each fiber because of excessive grain growth of SiC grains. Typically, similar to the case of the densification of monolithic SiC, the matrix layers were well sintered at 1750°C. The composite sintered for 1 h shows micropores in the inter-fiber areas (see Fig. 3(a)) because the spherical morphology of the fibers was maintained in the composite. Fiber morphology changed from a spherical shape in the 1-h-sintered composite (Fig. 4(a)) to a hexagonal shape in the 4-h-sintered composite (Fig. 4(c)) with increasing the sintering time, indicating the deformation of fibers at 1750°C under an applied pressure of 15 MPa. Therefore, the further densification observed in the 4-h-sintered composite (Fig. 2) was due to the inter-fiber densification induced by fiber deformation.

Fig. 4 shows that the grain size of the SiC grains (1.0~2.0 μm) in the fibers of the 4-h-sintered composite was larger than that of the SiC grains (0.5~1.0 μm) in the fibers of the 1-h-sintered composite. Hence, prolonged annealing at

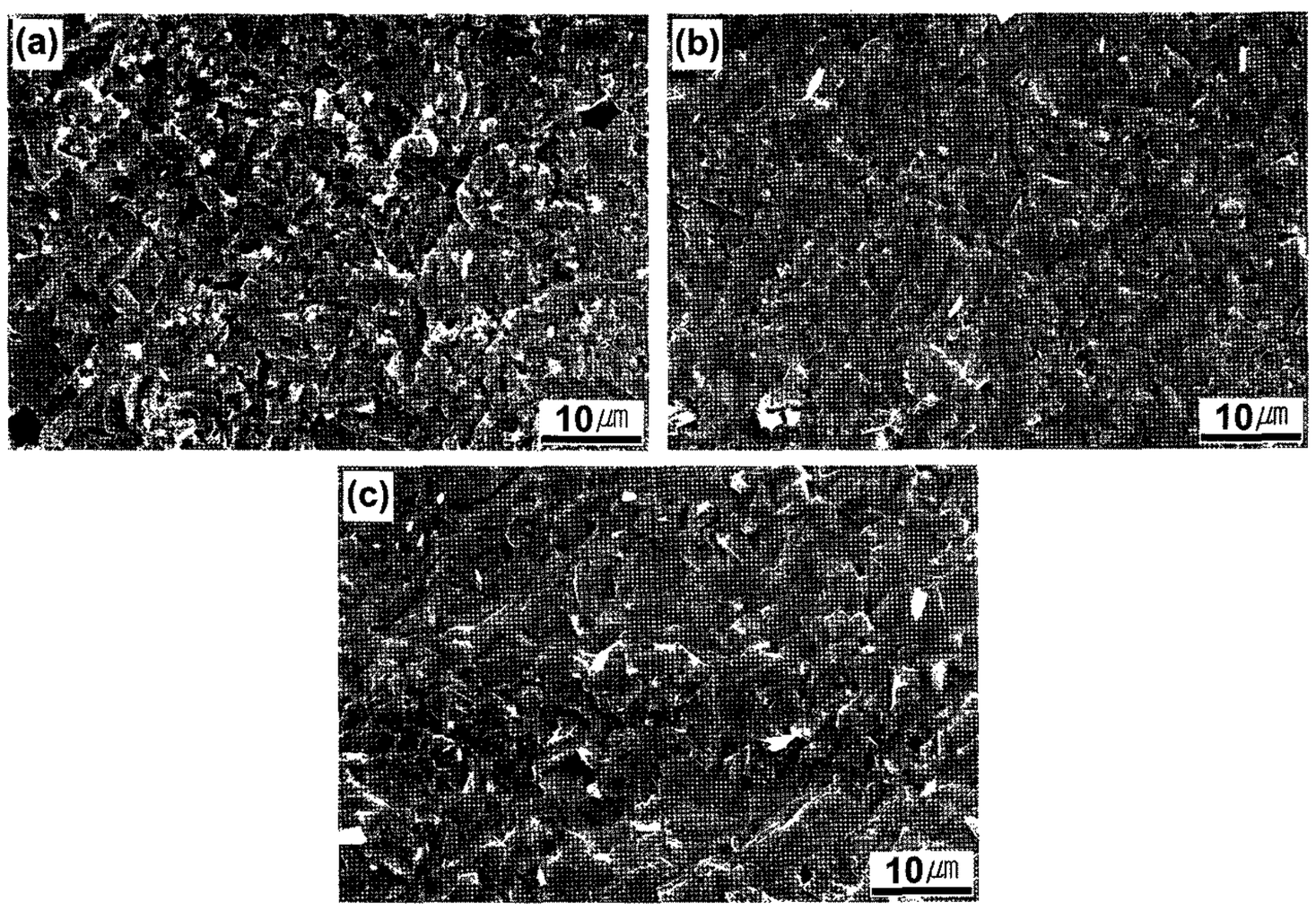

Fig. 3. Typical fracture surfaces of the SiC fiber-SiC matrix composites sintered for various times: (a) 1 h, (b) 2 h, and (c) 4 h.

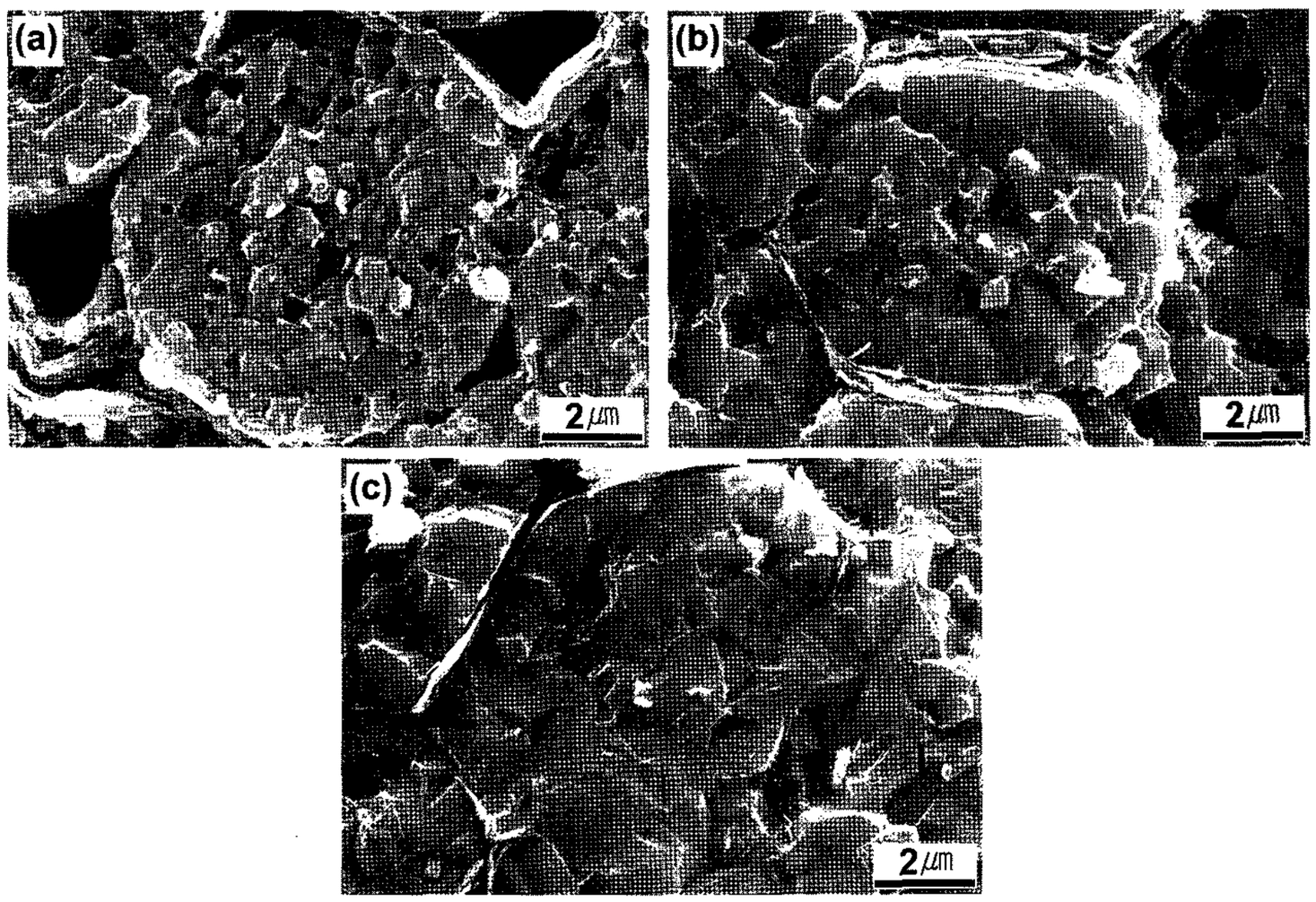

Fig. 4. Magnified SEM micrograph of the SiC fiber-SiC matrix composite sintered at 1750°C for (a) 1 h, (b) 2 h, and (c) 4 h.

1750°C increased both the density of the composite as well as the grain size of the SiC grains in the fibers.

Fig. 2 also shows the variations in flexural strength for each composite fabricated at different sintering times. The strength increased with increasing the sintering time. A maximum strength of ~401 MPa was obtained from the 4-h-sintered composite. The increase in strength after prolonging the sintering time was due to the increased densification in the 4-h-sintered specimen and the growth of the elongated grains to moderate sizes (5-8 μm) in the matrix (see Fig. 3(c)) of the 4-h-sintered specimen. The growth of the elongated grains with a moderate size in both the matrix and fibers generally led to increased strength and toughness in the SiC matrix composites.[18,19]

A flexural strength of ~280 MPa was reported in a 2D SiC fiber-SiC matrix composite fabricated by a CVI process when no pyrolytic carbon coating was applied.[4] The sintered density of the composite was 2.78 g/cm^3. Flexural strength values of 450-500 MPa were reported in SiC/SiC composites that were fabricated via slurry infiltration and stacking process.[16] The sintered densities of the composites ranged from 3.03 to 3.18 g/cm^3, depending on sintering time. The strength (~401 MPa) of the 1750°C-sintered SiC/SiC composite fabricated by the present process is in between the

CVI processed composites and the slurry infiltration and stacking processed composites. The sintered density of the composites fabricated here ranged from 3.03 to 3.06 g/cm^3. Those values are also in between both composites. These results suggest that the strength of SiC/SiC composites depends primarily on the sintered density of the composites and the absence of the degradation of the fibers during processing.

4. Conclusion

Dense (3.03~3.06 g/cm^3, 96.4~97.6% TD) SiC fiber reinforced SiC matrix composites were obtained via a reaction sintering route. The process involves infiltration of the SiC fiber fabric using a slurry consisting of Si and C precursors, stacking of the infiltrated fabric, cross-linking, pyrolysis, and (v) hot-pressing of the pyrolyzed composites. Typical flexural strength of the 4-h-sintered composite was ~400 MPa.

Acknowledgment

This study was supported by a grant from the Fundamental R&D Program for Core Technology of Materials funded by the Ministry of Commerce, Industry, and Energy, Republic of Korea.

REFERENCES

1. H. Ohnabe, S. Masaki, M. Onozuka, K. Miyahara, and T. Sasa, "Potential Application of Ceramic Matrix Composites to Aero-Engine Components," *Comp. Part A*, **30** [4] 489-96 (1999).
2. X. Luan, L. Cheng, Y. Xu, and L. Zhang, "Stressed Oxidation Behaviors of SiC Matrix Composites in Combustion Environments," *Mater. Lett.*, **61** [19-20] 4114-16 (2007).
3. R. Naslain, "Design, Preparation and Properties of Non-Oxide CMCs for Application in Engines and Nuclear Reactors: An Overview," *Comp. Sci. Tech.*, **64** [2] 155-70 (2004).
4. W. Yang, T. Noda, H. Araki, Y. Jinnan, and A. Kohyama, "Mechanical Properties of Several Advanced Tyranno-SA Fiber Reinforced CVI-SiC Matrix Composites," *Mater. Sci. Eng. A*, **345** [1-2] 28-35 (2003).
5. J. Y. Park, H. S. Hwang, W. J. Kim, J. I. Kim, J. H. Son, B. J. Oh, and D. J. Choi, "Fabrication and Characterization of SiC_f/SiC Composite by CVI using the Whiskering Process," *J. Nuclear Mater.*, **307-311** [2] 1227-31 (2002).
6. D. Jaglim, J. Binner, B. Vaidhyanathan, C. Prentice, B. Shatwell, and D. Grant, "Microwave Heated Chemical Vapor Infiltration: Densification Mechanism of SiC_f/SiC Composites," *J. Am. Ceram. Soc.*, **89** [9] 2710-17 (2006).
7. A. Kohyama, M. Kotani, Y. Katoh, T. Nakayasu, M. Sato, T. Yamamura, and K. Okamura, "High-Performance SiC/SiC Composites by Improved PIP Processing with New Precursor Polymers," *J. Nuclear Mater.*, **283-287**, 565-69 (2000).
8. M. Kotani, T. Inoue, A. Kohyama , Y. Katoh, and K. Okamura, "Effect of SiC Particle Dispersion on Microstructure and Mechanical Properties of Polymer-Derived SiC/SiC Composite," *Mater. Sci. Eng. A*, **357** [1-2] 376-85 (2003).
9. Y. Katoh, S. M. Dong, and A. Kohyama, "Thermo-Mechanical Properties and Microstructure of Silicon Carbide Composites Fabricated by Nano-Infiltrated Transient Eutectoid Process," *Fusion Eng. Design*, **61-62**, 723-31 (2002).
10. S. Dong, Y. Katoh, and A. Kohyama, "Preparation of SiC/SiC Composites by Hot Pressing, using Tyranno-SA fiber as Reinforcement," *J. Am. Ceram. Soc.*, **86** [1] 26-32 (2003).
11. M. S. Kim, Y.-W. Kim, K. S. Han, and J. H. Park, "Nicalon-Fiber-Reinforced SiC Composites via Infiltration(*in Korean*)," *J. Kor. Ceram. Soc.*, **30** [12] 993-98 (1993).
12. Y.-W. Kim, J. G. Lee, M. S. Kim, and J. H. Park, "Effect of Multilayer Coating on Mechanical Properties of Nicalon-Fiber-Reinforced Silicon Carbide Composites," *J. Mater. Sci.*, **31** 335-38 (1996).
13. Y.-W. Kim, Y. S. Chun, J. Y. Park, W. S. Ryu, and D. S. Park, "Process for Dense 2D SiC Fiber-SiC Matrix Composites," *Met. Mater. Int.*, **13** [3] 197-200 (2007).
14. K. Y. Lim, D. H. Jang, Y.-W. Kim, J. Y. Park, and D. S. Park, "Effect of Processing Parameters on the Densification and Strength of 2D SiC Fiber-SiC Matrix Composites Fabricated by Slurry Infiltration and Stacking Process," *J. Kor. Ceram. Soc.*, **44** [7] 349-53 (2007).
15. J. H. Eom, D. H. Jang, Y.-W. Kim, I. H. Song, and H. D. Kim, "Low Temperature Processing of Porous Silicon Carbide Ceramics by Carbothermal Reduction(*in Korean*)," *J. Kor. Ceram. Soc.*, **43** [9] 552-57 (2006).
16. Y.-W. Kim, S. H. Kim, I. H. Song, H. D. Kim, and C. B. Park, "Fabrication of Open-Cell, Microcellular Silicon Carbide Ceramics by Carbothermal Reduction," *J. Am. Ceram. Soc.*, **88** [10] 2949-51 (2005).
17. S. M. Lee, T. W. Kim, H. J. Lim, C. Kim, Y.-W. Kim, and K. S. Lee, "Mechanical Properties and Contact Damages of Nanostructured Silicon Carbide Ceramics," *J. Ceram. Soc. Jpn.*, **115** [5] 304-09 (2007).
18. Y.-W. Kim, M. Mitomo, and H. Hirotsuru, "Grain Growth and Fracture Toughness of Fine-Grained Silicon Carbide Ceramics," *J. Am. Ceram. Soc.*, **78** [11] 3145-48 (1995).
19. Y.-W. Kim, Y. S. Chun, T. Nishimura, M. Mitomo, and Y. H. Lee, "High-Temperature Strength of Silicon Carbide Ceramics Sintered with Rare-Earth Oxide and Aluminum Nitride," *Acta Mater.*, **55** [2] 727-36 (2007).